MEASUREMENT OF THE DIELECTRIC CONSTANT OF TRIGLYCINE SULFATE AT MICROWAVE FREQUENCIES

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THESIS

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ABSTRACT

This thesis describes the construction and operation of a microwave system used to measure very high dielectric constants over wide frequency and temperature ranges. Measurements of the dielectric constant of the ferroelectric triglycine sulfate are reported over a frequency range of 4.0 to 12.0 GHz. The results agree fairly well with those reported by other investigators. Sources of errors and possible improvements in this experiment are discussed.

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I. INTRODUCTION

Ferroelectric materials have properties that make them attractive in device applications. Research is being done in many laboratories to adapt ferroelectrics to infrared detectors, parametric amplifiers, and a broad range of display and memory devices. The property of interest in this study is the very high dielectric constant, which is field and temperature dependent. For many materials the dielectric constant remains high well beyond the microwave range. For device application good dielectric data is needed, however, there is little reliable information available about any material with high dielectric constants in the microwave ranges.

The purpose of this project then, was to measure the dielectric constant for the ferroelectric triglycine sulfate, abbreviated TGS, over the frequency range of 4.0 to 12.0 GHz and a temperature range of 20 to 80^{o}C. TGS is a second order ferroelectric with a transition temperature of approximately 49^{o}C.

The properties of TGS have previously been investigated with the earliest comprehensive reports of measurements of its dielectric constant appearing in 1958. [Ref. 1.] Since then numerous articles have appeared reporting on measurements of its dielectric constant at various frequencies and over various temperature ranges. References 2 and 3 are representative. Very few of these reports agree and the results reported are generally considered unreliable Reference 4 contains one of the most comprehensive discussions of the properties of TGS including its thermodynamic properties.

A variety of experimental techniques are available for performing the measurement of the dielectric constant of a substance. A thorough

discussion of some of these techniques is found in reference 5. The method used in this experiment is reflection with a shortcircuited coaxial line. A small sample is placed between the end of the center conductor and a metal plate placed across the end of the outer conductor of the coaxial line. The sample itself, thus becomes a part of the short circuit. Sandy, [Ref. 5], has shown that for cylindrical samples that are small compared to the coaxial line, the field is very nearly uniform across the sample. The behavior of the sample is approximately that of a capacitance with the dimensions of the sample and a complex dielectric constant. The motivation for using a coaxial system is that it is possible to have a single system that can be used over a wide frequency range.

The sample holder must be calibrated using materials of known dielectric constant, so the accuracy of the calculated dielectric constant for TGS as determined from the experimentally measured parameters is affected by the accuracy with which their dielectric constants are known. Lithium flouride, sodium chloride, and teflon were used as standards by Porter, [Ref. 6], utilizing the dielectric constant values reported for these materials by von Hippel, [Ref. 7]. However, these values were considered not reliable over the frequency range being used in this experiment, so rochelle salt was taken as the primary standard. Values of its dielectric constant as reported by Sandy, [Ref. 5], were utilized. The calibration procedure used is discussed in Chapter II.

The basic procedures used in making the measurements on TGS were originally utilized by Sandy [Ref. 5], modified by Porter, [Ref. 6], and further modified for this experiment as discussed in Chapter II. The TGS samples used in this experiment were grown and prepared in our laboratory. Details of the sample preparation are described in Chapter II. A computer program was used to reduce the experimental data. Chapter III contains

a discussion of how the data was evaluated. The results and conclusions are discussed in Chapter IV.

II. EXPERIMENTAL PROCEDURES

A. EQUIPMENT

Measurements of the dielectric constant of TGS were made over a frequency range of 4.0 GHz to 12.0 GHz and a temperature range of 20°C to 80°C.

A diagram of the instrumentation used for making the measurements is shown in Figure II-1. This system is similar in many respects to that used by Porter, [Ref. 6], however, a sweep generator replaces the klystron used by Porter and the sample holder was modified. The signal was fed from the sweep generator via a coaxial slotted line with a moveable probe and a bolometer in a tunable mount. A slide-wire rheostat was mounted on the carriage of the slotted line to provide X-axis drive for an X-Y recorder. The output from the bolometer was fed to a Hewlett-Packard Model 415 B Standing Wave Indicator, and then to the Y-axis drive of the X-Y recorder. The curve thus generated was used to locate voltage minima and to determine phase changes and wavelength.

Next in line was a pair of directional coupliers. Attached to the directional coupliers were two Crystal Detectors, Model HP 423A (Neg.). By using these detectors, which have a very flat frequency response, it was possible to monitor the reflected signal and also to maintain constant power output over the full sweep generator range. (See Figure II-1.) The ratio meter's output was used to drive the Y-axis of a second X-Y recorder. The X-axis signal of this recorder was supplied by a voltage from the sweep generator that was proportional to frequency, thus producing a curve of reflection coefficient as a function of frequency. The evaluation of these plots is discussed below.

The sample holder used is shown in Figure II-2. Because of the

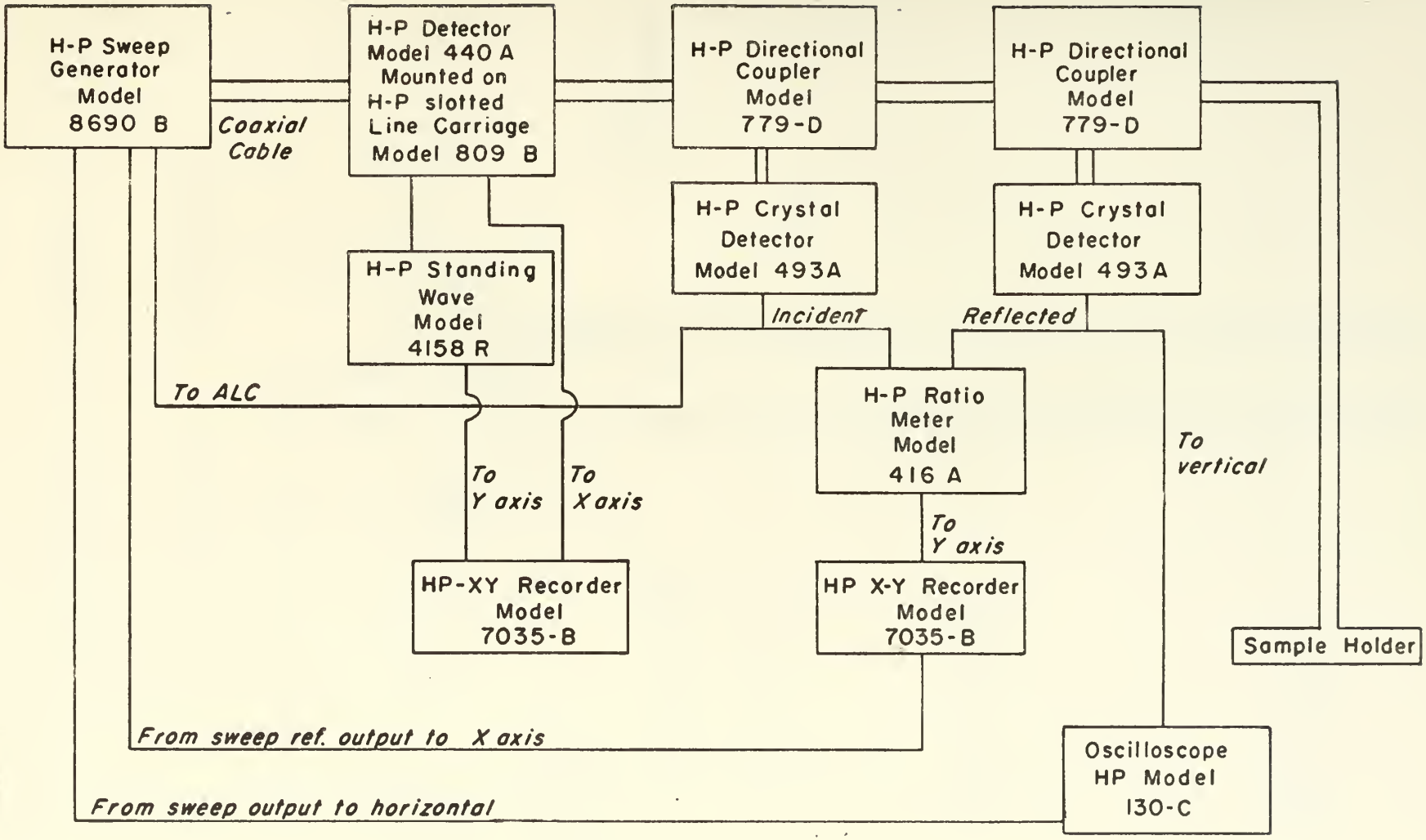

Figure II-1. Experimental Configuration

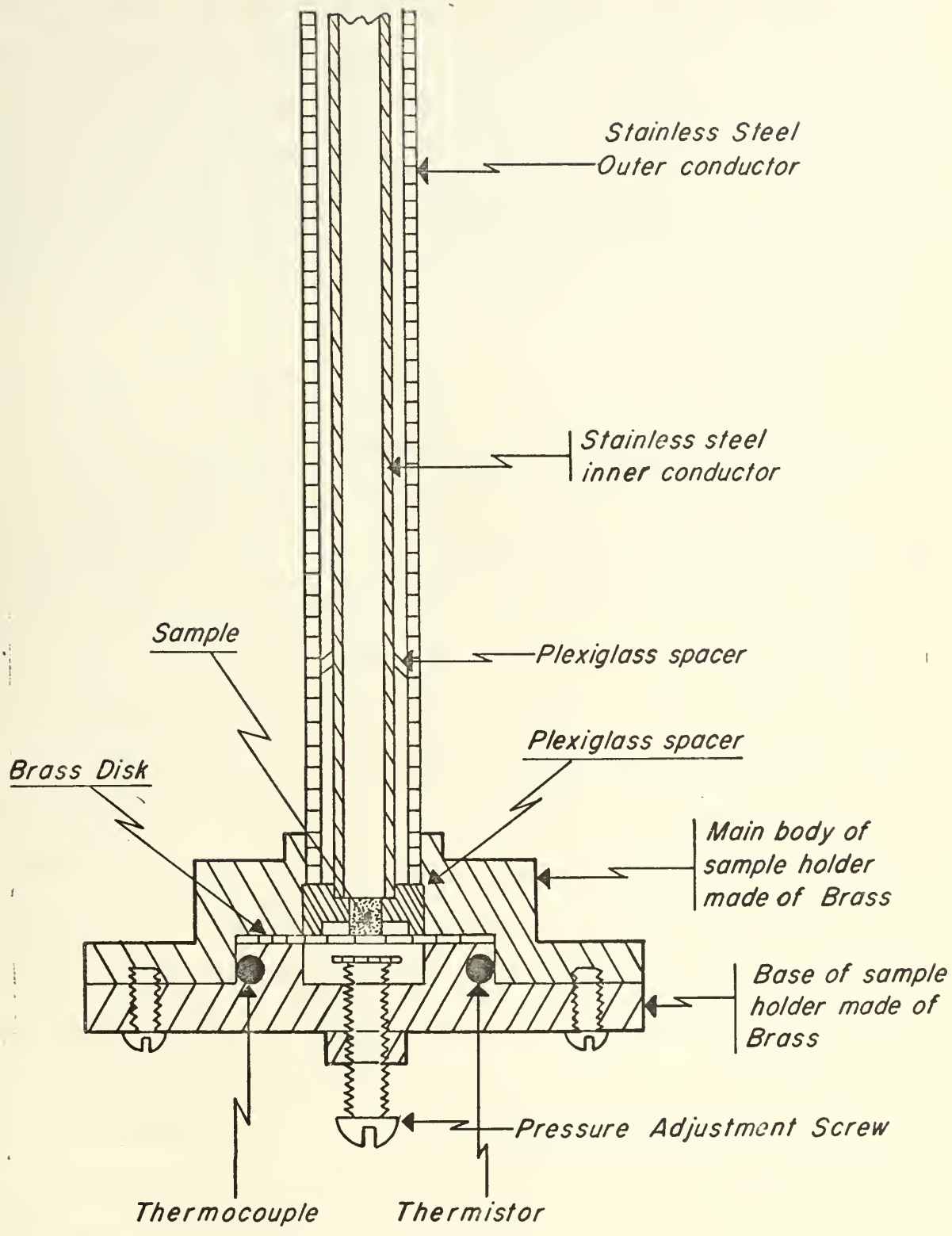

Figure II-2. Sample Holder

piezoelectric characteristic of TGS and rochelle salt, the sample holder was equipped with an adjustable screw on the bottom so that the pressure exerted on the sample by the brass shorting plate could be varied. The mass of the sample holder is small so that it can come quickly to thermal equilibrium as the temperature is changed. A thermistor and thermocouple were mounted on the holder to provide a means for monitoring the temperature of the sample.

B. TEMPERATURE CONTROL

Since the dielectric constant of TGS is temperature dependent, it was necessary to measure the dielectric constant of the sample over a wide temperature range. The TGS measurements were made over a temperature range of 20°C to 80°C. Calibration data for rochelle salt was taken over the range -50°C to +50°C. Figure II-3 shows the system used to control the temperature of the sample.

The sample holder with the sample in place was mounted in an insulated glass tube and temperature controlled air flowed through the tube, maintaining the sample at the desired temperature. For the TGS measurements, air from a laboratory outlet was fed into a Varian Variable Temperature System, Model V-4257. The rate of air flow could be adjusted by means of the flow meter on the Model V-4257. In the gas flow, two platinum temperature sensors were mounted. These sensors are in the loop that controls the current flowing to the heating coils.

The system used for measuring the temperature is also depicted in the figure. A thermistor was mounted on the side of the sample holder near the sample. The thermistor is one arm of an a.c. wheatstone bridge. The error signal from the bridge is detected by a Lock-in-Amplifier whose output is displayed on a strip chart recorder. By

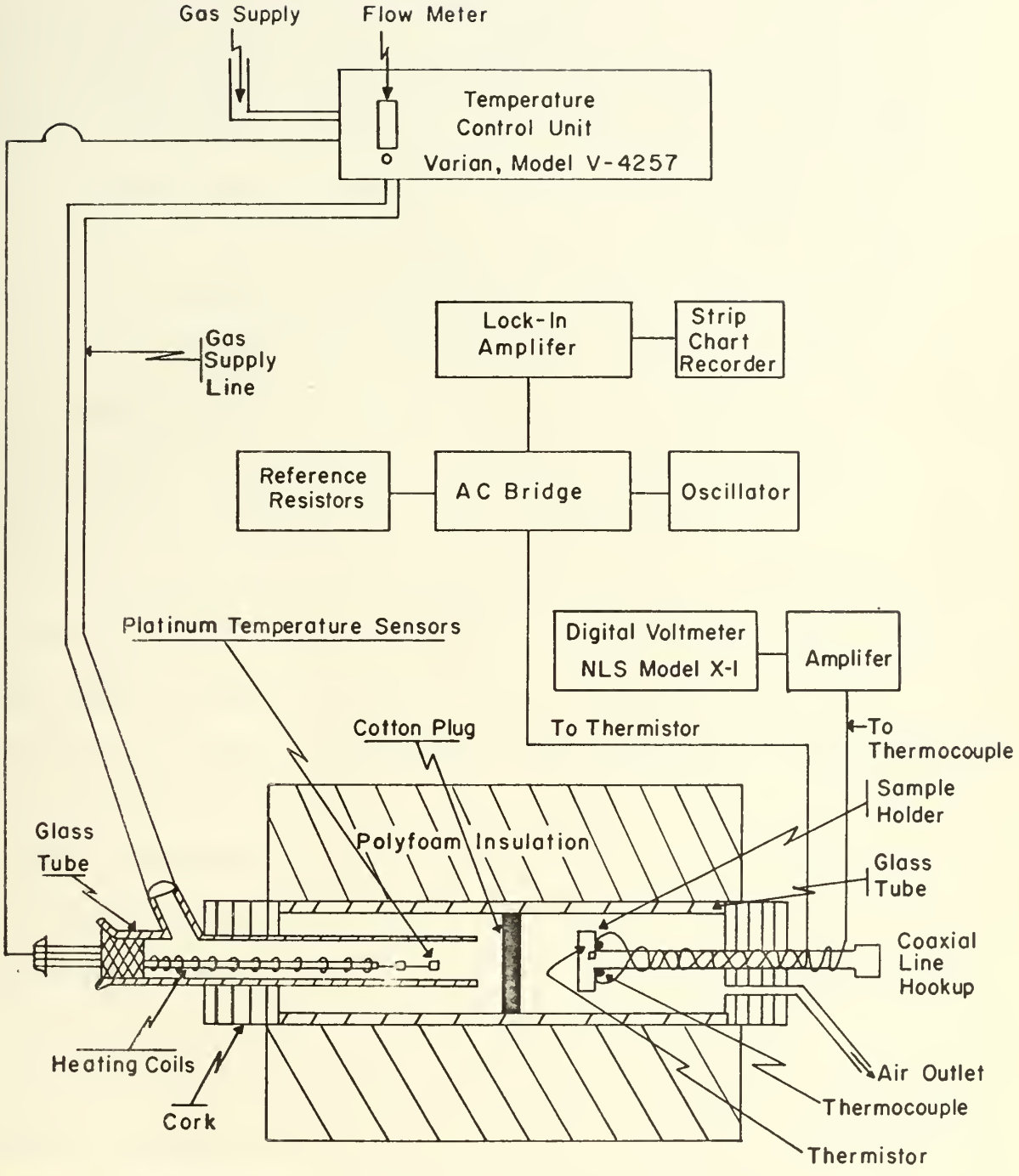

Figure II-3. Temperature Control System

changing the settings on the reference resistors, the bridge could be balanced. This system may be used to measure the temperature of the sample very accurately, however, in this case, it was used to continuously monitor the thermal equilibrium of the sample.

For actually measuring the temperature of the sample, a copper-constantin thermocouple was mounted in the sample holder. The voltage from the thermocouple was originally read on a Keithley Model 149 Milli-Microvoltmeter. Later a NLS Series X-1 Digital Voltmeter was utilized when this piece of equipment became available. The wires of the thermocouple were wound tightly around the microwave coaxial conductor leading to the sample holder in an effort to cut down on heat losses due to conduction down the wires. It is estimated that the temperature measurements obtained with this system were accurate to about $\pm$ 0.3°C.

For the low temperature work using rochelle salt, nitrogen gas was used instead of air to prevent the formation of ice in the lines leading to the sample. Liquid nitrogen was used to cool the gas just prior to its entrance into the tube containing the heating coils where it was warmed up to the desired temperature.

C. SAMPLE PREPARATION

Cylindrical samples were prepared by approximately the same method as described by Sandy. [Ref. 5]. This method entails cutting the sample to approximate size, gluing them to a small steel rod, which is in turn mounted in the chuck of a jeweler's drill press. For water insoluble materials, sandpaper can be used to wear the sample down to the desired diameter. If the material is water soluble like TGS and rochelle salt, a moist cloth held tightly against the sample while it is being turned at high speed in the drill press, will smooth it down satisfactorily. A sharp razor blade can then be used to cut the sample to the approximate length.

A metal jig of proper thickness and holes in it of proper diameter was constructed to help insure all samples were of the same diameter and length. The sample was put in the jig and the ends smoothed down until the sample was the correct length. The brass sample, which was used as one of the standards, was prepared on a metal lathe. The samples used had a diameter and length of 0.19 ± 0.005 centimeters.

The dielectric constant of TGS was measured along the monoclinic b axis, so it was important that samples prepared from a crystal were oriented properly with respect to this axis. This was not much of a problem since TGS crystals have a cleavage plane perpendicular to the b axis. The crystal was cleaved by simply placing a razor blade across the crystal and taping lightly until it cleaved. If the crystal broke with something other than a smooth edge, you could safely say that this was not the desired cleavage plane. Jona and Shirane in reference 4 discuss the reference system of coordinates in a TGS crystal and how one can go about determining the b axis.

The TGS crystals from which the samples were made were grown in our laboratory from a water solution prepared by reacting an aqueous glycine solution with the proper amount of sulfuric acid. One mole of glycine, that is 75.07 grams, was dissolved in 300 ml of warm water, then 18.57 ml (1/3 mole) of 96% sulfuric acid was added. The formula for TGS is $(NH_2CH_2COOH)_3H_2SO_4$. After standing for a few days exposed to the air at room temperature, TGS crystals start to precipitate out of solution. Once the solvent has almost all evaporated, the remaining solvent is poured off. Additional water is then used to redissolve the crystals, and again the solvent is evaporated off. The procedure can be repeated several times to purify the TGS crystals.

Some of the crystals grown in this manner contained flaws. This

was evident when trying to prepare a sample. Some of the crystals would not turn down smoothly, some parts wearing away faster than others. Because of this difficulty, quite a few attempts at sample preparation ended in failure. Once a sample was finished without any flaws, the ends were coated with silver paint to insure good contact when the sample was installed in the sample holder.

The samples of rochelle salt were prepared from a Y-cut plate of the salt. In order to locate the ferroelectric axis, cuts were made from the main plate at various orientations. These small samples were smoothed down, the surfaces coated with silver paint, and then placed in a capacitance bridge. The samples were cooled and their dielectric behavior observed. The correct orientation was found by checking to see which of the samples had an anomaly at the transition temperature.

D. MEASUREMENT PROCEDURE AND DATA COLLECTION

Prior to making any measurements, the ratio meter had to be calibrated. To accomplish this, an open line was placed where the sample holder normally was attached to the coaxial system. The sweep generator was then set to the highest frequency being used. Next, the power setting on the sweep generator was adjusted so that 40 millivolts peak to peak voltage was observed on the oscilloscope. (See Figure II-1 for system configuration.) The "set to full scale" knob on the ratio meter is then set for 100% reflection. Next, the Y-axis of the X-Y recorder being used to record the reflection coefficient is set to a maximum position. The X-axis is adjusted so that the entire frequency range being swept will just fill the paper. A plot of the reflection coefficient is then made over the frequency range. The open line is then replaced with a short and another plot made. These two plots are then averaged, giving us a

calibration of 100% reflection coefficient over the frequency range being swept. This same procedure is followed for 90%, 80%, etc. The resulting set of curves is then used as a standard against which reflection coefficient curves for the samples can be compared. A typical calibration curve is shown in Figure II-4.

The information needed from the measurements consists of the reflection coefficients of the TGS sample and calibration standards at various frequencies and various temperatures, as well as the phase shift at these same frequencies and temperatures. In order to calibrate the sample holder, these parameters were measured for a brass sample, the empty sample holder, and a rochelle salt sample. Then by comparing the measured values of the reflection coefficient and the phase shift for TGS to those obtained from the calibration samples, the dielectric constant for TGS could be determined. Chapter III contains a discussion of how the dielectric constant was evaluated from these parameters.

To make a measurement, a sample was placed in the sample holder and the holder placed on the line as shown in Figure II-1. Just prior to this, the sweep generator must be set to the highest frequency being used and, with an open line, the ratio meter set to read 100% reflection. With the sample holder in place, and the sweep generator operating in the continuous wave mode, the screw on the bottom of the sample holder must be adjusted to insure the proper pressure is being applied to the sample. Both TGS and rochelle salt are sensitive to pressure, that is, they are piezoelectric. Because of this effect, variations in pressure will give different readings on the ratio meter. When increasing the pressure on the sample by tightening the screw, the deflection of the needle on the ratio meter is observed. A plateau will be reached where with additional pressure, very little deflection of the ratio meter's needle will occur. (This effect

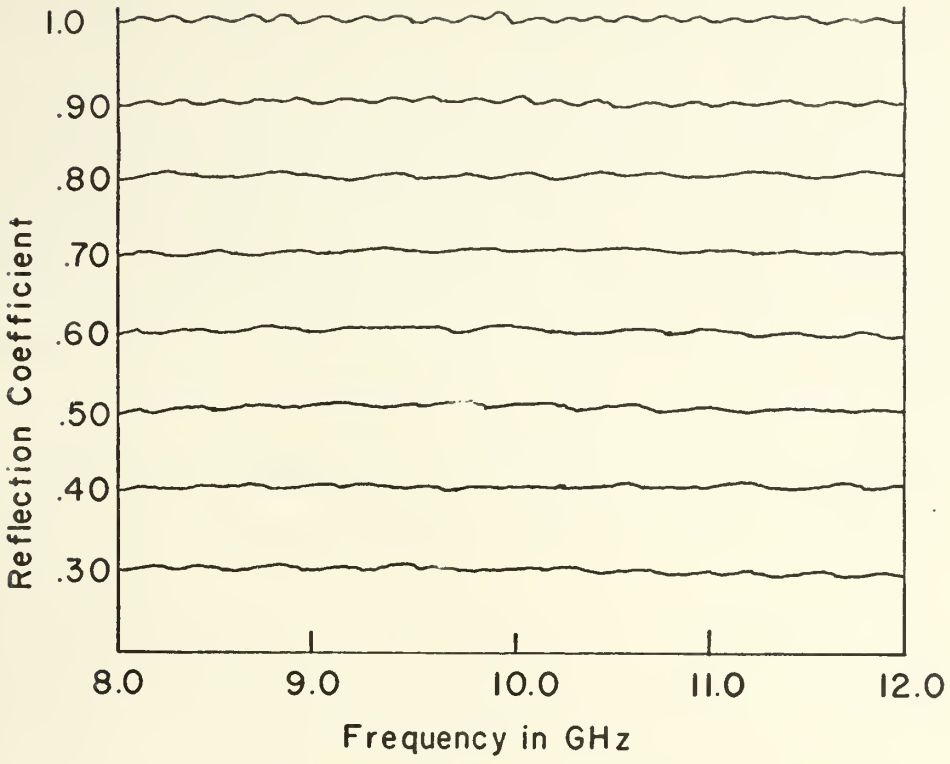

Figure II-4. Typical Calibration Curve

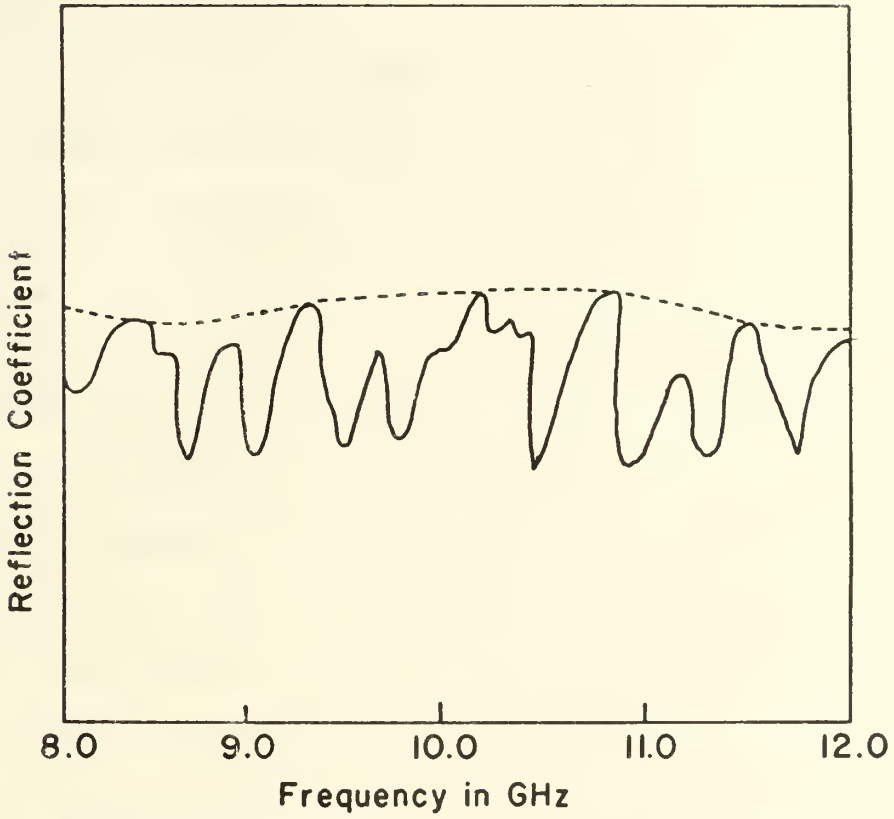

Figure II-5. Typical plot of refection co-efficient vs. frequency

is described by Sandy. [Ref. 5]) When the plateau was reached, this was then taken as the proper pressure setting. The sample holder was then placed in the insulated glass tube and the temperature control equipment hooked up. The sweep generator was then swept over the desired frequencies and the reflection coefficient was plotted by one of the X-Y recorders. A typical plot produced is shown in Figure II-5. The peaks and valleys on the plot are apparently caused by reasonances within the system. The height of the curve varies with temperature and frequency as does the position of the peaks and valleys. These resonances complicate data reduction, which is discussed below.

Before making a measurement, the system has to be in thermal equilibrium. It generally took about ten minutes to establish equilibrium each time the temperature was changed. The temperature was measured just prior to taking a reflection coefficient reading.

After making the reflection coefficient measurement, and holding the temperature constant, the sweep generator was switched to the continuous wave mode and set on a frequency at which phase information was desired. A plot was then made, at this frequency, of the standing wave pattern using the slotted line and X-Y recorder. Standing wave plots were made for the calibration samples at this same frequency. For TGS this procedure had to be repeated at each temperature and frequency of interest. This was not the case for the brass sample and the empty sample holder. It was determined that the parameters for these two samples were independent of temperature. The positions of the voltage minima from the standing wave plot for each sample were compared to the minima positions for the brass sample. In this way the phase shift was determined for each frequency and temperature of interest.

E. DATA REDUCTION

To obtain useful information from the measurements that was not being obscured by the shifting peaks and valleys as illustrated in Figure II-5, two different procedures were tried. The first was an averaging method. In this method, the peaks on the reflection coefficient versus frequency curves were connected by a smooth line as shown by the dashed curve on Figure II-5. The reflection coefficient was then read from the dashed line at a frequency of interest. The phase data was then taken at this same frequency.

Another method used is to designate prominent peaks on the reflection coefficient curve with a number, then follow this same peak over the temperature range being considered. Figure II-6 illustrates this method. The lower curve in the figure shows the reflection coefficient versus frequency for the TGS sample at two different temperatures. The position of the numbered peaks can be seen to change as the temperature is changed. The reflection coefficient is always read for peak position. This means that each reading will be at a slightly different frequency. The TGS phase information was taken at the frequency at which the peak occurred. Then, this in turn makes it necessary to know the position of the voltage minima in the standing wave pattern for the brass sample at many different frequencies. This was accomplished by making a plot of the standing wave pattern for the brass sample at each frequency of interest. The phase shift was then determined by comparing the minima positions for the brass sample against the positions of the minima for the TGS sample.

The peak identification method does introduce some difficulty. The upper curve shown in Figure II-6 is typical of the reflection coefficient versus frequency plot for any of the calibration samples. Matching the peaks in the calibration curve with the corresponding peaks for the TGS

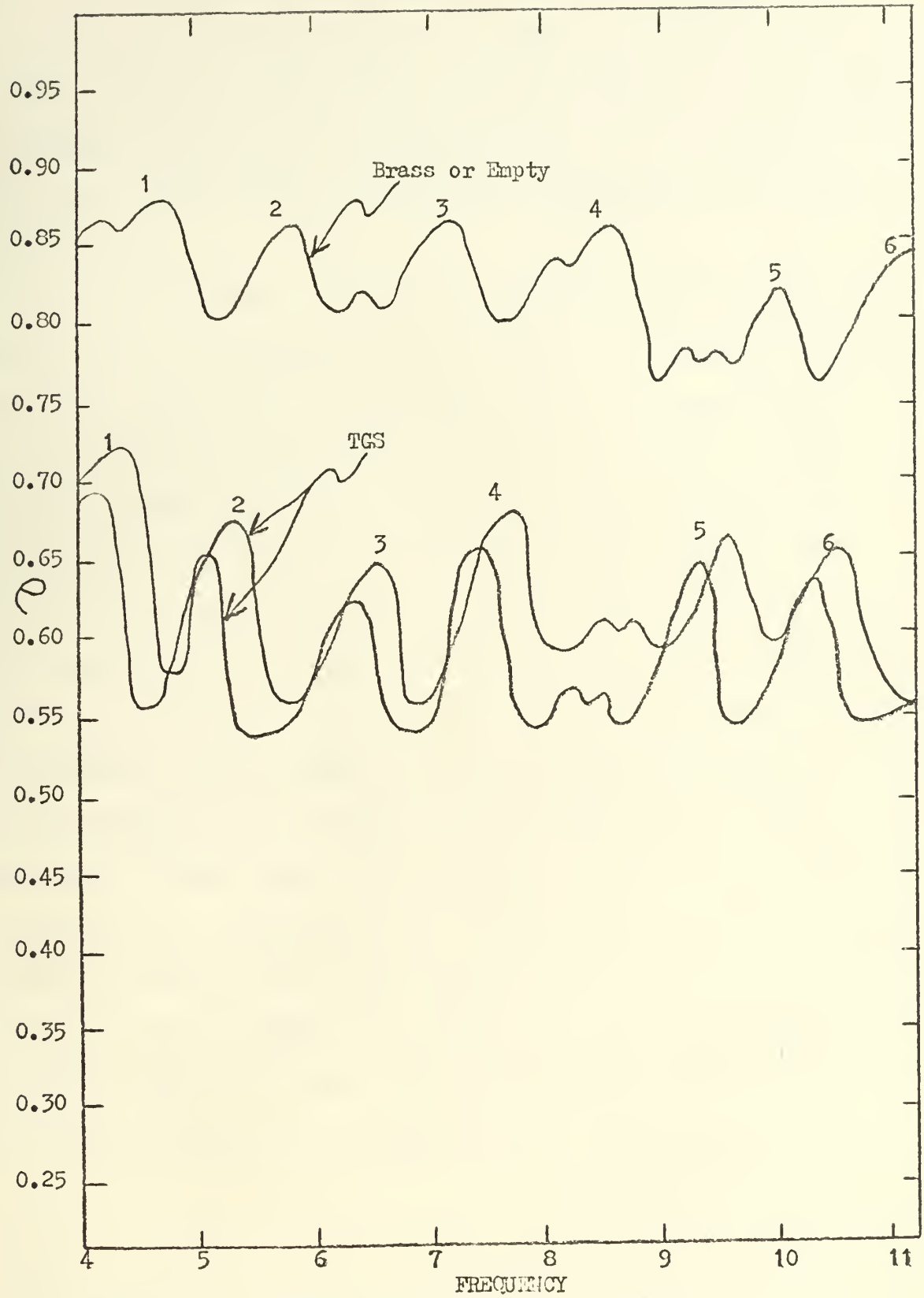

FIGURE II-6 REFLECTION COEFFICIENT vs FREQUENCY

curve is not always easy. Some peaks that show up in the brass curve or in the empty sample holder curve may not appear in the TGS curve or in the rochelle salt curve. The converse is also true. Thus, using this method, it is important that only prominent peaks common to all curves be selected as the ones at which to read the reflection coefficient and for measuring the phase shift.

It is estimated that the reflection coefficient could be read with an accuracy of $\pm$ 1% and the phase shift to about $\pm$ 2%. During the calibration work using rochelle salt, it was observed that it can easily lose the waters of hydration it contains in its molecular structure. At low temperatures when dry nitrogen was being used in the temperature control device, and at high temperatures when warm air was being used, after several hours of use, the originally clear sample would change to a milky white appearance and the sample would crumble easily. Before this happened, the sample had to be discarded and a new one used or considerable error could be introduced. The TGS samples did not have this problem, however, if the sample contained flaws in it which were not visible to the naked eye, the data taken for this sample could be slightly different than that taken for a sample that was supposedly perfect. These factors undoubtedly contributed to some uncertainty in the values obtained for the dielectric constant for TGS and also lead to difficulty in duplicating values using different but supposedly similar TGS samples. The results obtained from the above discussed measurements are presented in Chapter IV.

III. MATHEMATICAL THEORY AND DATA EVALUATION

The complex dielectric constant of TGS is calculated from the measured reflection coefficient and the phase of the standing wave pattern. The complex reflection coefficient is defined as:

$$\rho^* = \rho\left[\cos\ (4\pi x/\lambda) + i \sin\ (4\pi x/\lambda)\right] \tag{1}$$

where ρ is the reflection coefficient, λ the wavelength, and x the shift of a standing wave minimum from a reference plane. Then the complex admittance of the microwave line is given by:

$$Y^* = Y_0^*\ (1 + \rho^*)\ /\ (1 - \rho^*) \tag{2}$$

This relationship is discussed by Sandy in Reference 5. The characteristic admittance, Y_0^*, cancells out in subsequent equations and hence will be dropped from this point on.

The admittance of the sample is given by the relationship

$$Y_s^* = i\omega C^* = i\omega \mathcal{E}_s^* a/\ell \tag{3}$$

where ω is the angular frequency, $\mathcal{E}_s^*$ is the sample dielectric constant, a is the area and ℓ the length of the sample. The most general relationship between the sample and the line admittance is given by

$$Y^* = (A + BY_s^*)\ /\ (1 + CY_s^*) \tag{4}$$

where A, B, and C, are complex constants which are characteristic of the line and are a function of frequency. [Ref. 5 and 8.] By using equations 1 through 4, $\mathcal{E}^*$ can be related to ρ and x, provided A, B, and C have known values.

A, B, and C are obtained using samples of known dielectric constant to calibrate the system. Any set of three materials will suffice. We chose brass, the empty sample holder, and a single dielectric material for ease of computation. For these samples the following assumptions were made:

Brass: Y_b^* is very large and real. The zero of the phase shift is taken

at the brass standing wave minima. Air, that is the empty sample holder: Y^*_a is very small. From equation (4) it is then found that $Y^*_a = A$. Calibration dielectric: Y^*_c is given by equation (3), where $\mathcal{E}^*_c$ is known.

After a few algebraic steps the following expressions were obtained for the calibration constants:

$$A = (1 + \rho^*_a) \;/\; (1 - \rho^*_a) \tag{5}$$

$$B = C(1 + \rho_b) \;/\; (1 - \rho_b) \tag{6}$$

$$C = \frac{1}{i\omega \mathcal{E}^*_c D_c} \cdot \frac{(1 + \rho^*_c) - A(1 - \rho^*_c)}{[(1 + \rho_b) \;/\; (1 - \rho_b)]\,(1 - \rho^*_c) - (1 + \rho^*_c)} \tag{7}$$

where $D = a/\ell$. The subscripts a, b and c, refer to air, brass, and the calibration dielectric respectively. The samples dielectric constant is given by

$$\mathcal{E}^*_s = \frac{1}{i\omega_s D_s} \cdot \frac{(1 + \rho^*_s) - A(1 - \rho^*_s)}{B(1 - \rho^*_s) - C(1 + \rho^*_s)} \tag{8}$$

If a new parameter, $\Omega = (1 + \rho^*) \;/\; (1 - \rho^*)$ is defined, using equations (5) through (7) equation (8) can be rewritten in the following form:

$$\mathcal{E}^*_s = \mathcal{E}^*_c \;\frac{D_c}{D_s} \cdot \frac{\omega_c}{\omega_s} \cdot \frac{(\Omega_s - \Omega_a)(\Omega_b - \Omega_a)}{(\Omega_b - \Omega_a)(\Omega_c - \Omega_a)} \tag{9}$$

Note that the correction ω_c/ω_s is included. This is due to the fact that in certain circumstances the reflection coefficients may be taken at slightly different frequencies.

One further equation for $\mathcal{E}^*_s$ was derived using equation (9). It is:

$$\mathcal{E}^*_s = \mathcal{E}^*_c \;\frac{D_c}{D_s} \cdot \frac{\omega_c}{\omega_s} \cdot \frac{(1 - \rho^*_c)(\rho^*_s - \rho^*_a)(\rho_b - \rho^*_a)}{(1 - \rho^*_a)(\rho_b - \rho^*_s)(\rho^*_c - \rho^*_a)} \tag{10}$$

Any one of these equations, that is (8), (9), and (10), may be used to evaluate the sample dielectric constant from the data. The choice of which one to use is a matter of taste, dictated somewhat by the way calibration information is obtained. For example, equation (8) allows one to use a predetermined set of A, B and C values, then by interpolation $\mathcal{E}^*_s$ could be found at the frequency ω_s. On the other hand, by using equation (10), $\mathcal{E}^*_s$

is obtained more directly from the data since no calibration constants are required, but ρ_b and ρ_a^* need to be nearly frequency independent or all data must be taken at the same frequency. Both equations (8) and (10) were used with the final results being obtained from the latter. The method for calculating the dielectric constant from the measured parameters is discussed below.

The calculations were done on a Hewlett-Packard Model 9100A desk computer. A listing of the computer program is presented below. The list shows the program step, the computation key that is used for that step, and the computer code for the key. The computer code is given so that a program check can be made easily using the print out on the computer.

Step	Key	Code	Step	Key	Code	Step	Key	Code
00	clear	20	22	↑	27	44	1	01
01	stop	41	23	sin x	70	45	-	34
02	x	36	24	↑	27	46	b	14
03	y → ()	40	25	stop	41	47	y⇄x	30
04	9	11	26	x	36	48	to polar	62
05	↑	27	27	y → ()	40	49	ln x	65
06	stop	41	28	d	17	4a	acc -	63
07	x	36	29	roll ↑	22	4b	stop	41
08	y → ()	40	2a	cos x	73	4c	roll ↑	22
09	d	17	2b	x	36	4d	-	34
0a	stop	41	2c	y → ()	40	50	b	14
0b	roll ↑	22	2d	c	16	51	ch sgn	32
0c	x	36	30	1	01	52	y⇄x	30
0d	↓	25	31	-	34	53	to polar	12
10	↑	27	32	↓	25	54	ln x	65
11	sin x	70	33	to polar	62	55	acc +	60
12	↑	27	34	ln x	65	56	stop	41
13	stop	41	35	acc +	60	57	y⇄()	24
14	x	36	36	↓	25	58	9	11
15	y → ()	40	37	b	14	59	x	36
16	b	14	38	-	34	5a	↓	25
17	roll ↑	22	39	c	16	5b	↑	25
18	cos x	73	3a	↑	27	5c	cos x	73
19	x	36	3b	a	13	5d	↑	27
1a	y → ()	40	3c	-	34	60	stop	41
1b	a	13	3d	roll ↓	31	61	x	36
1c	stop	41	40	to polar	62	62	y → ()	40
1d	d	17	41	ln x	65	63	c	16
20	x	36	42	acc -	63	64	roll ↑	22
21	↓	25	43	↓	25	65	sin x	70

Step	Key	Code	Step	Key	Code	Step	Key	Code
66	x	36	74	acc +	60	81	to polar	62
67	y → ()	40	75	stop	41	82	ln x	65
68	d	17	76	c	16	83	acc +	60
69	b	14	77	-	34	84	Rcl	61
6a	-	34	78	d	17	85	e^x	74
6b	c	16	79	ch sgn	32	86	to rect	66
6c	↑	27	7a	y ⇄ x	30	87	↑	27
6d	a	13	7b	to polar	62	88	↓	25
70	-	34	7c	ln x	65	89	÷	
71	↓	25	7d	acc -	63	8a	print	45
72	to polar	62	80	Rcl	61	8b	end	46
73	ln x	65						

Each time a stop appears in the program, the calculation stops, then, a new piece of data is entered by the operator and the continue key on the keyboard is depressed to continue the calculation. The data entered at the stops, and the register into which it is entered are shown below.

Stop	X	Y
1	$4\pi/c$	ν_s
2	ν_c	
3	x_e	
4	ρ_e	
5		x_c
6	ρ_c	
7	ρ_b	
8	x_s	
9	ρ_s	
10		ρ_b
11	ε_{c1}	ε_{c2}

The frequencies, ν, are read in GHz. $4\pi/c = 0.41916$. The values x are the difference in position of the brass and sample minima in centimeters.

The output appears on the X, Y and Z registers of the computer and on the print tape. The values given in the three registers are: $X = \varepsilon_{s1}$, $Y = \text{Tan } \delta$, $Z = \varepsilon_{s2}$. These results need to be multiplied by ν_c/ν_s if the two frequencies were not the same.

IV. RESULTS and CONCLUSIONS

A. GENERAL REMARKS

The calculated results for the dielectric constant of TGS, both real and imaginary parts, as determined from the measured parameters, are shown in Figures IV-2 through IV-7. The values for the loss tangent are shown in Figures IV-8 through IV-10. Plots of dielectric constant versus frequency, at constant temperature, are shown in Figure IV-11.

The results shown are in general agreement with the results reported by Lurio and Stern in reference 9. They report that both $\mathcal{E}_1$ and $\mathcal{E}_2$ for TGS are around 6000 for a frequency of 1.0 kc, decreasing to about 2000 at 1.0 GHz, and then gradually decreasing to 1000 at 2.0 GHz. These values are taken at the transition temperature. Our results clearly exhibit the characteristic behavior of TGS in the vicinity of the transition temperature. For low frequencies, near 49°C, the dielectric constant rises to a high value, for example, $\mathcal{E}_1 \sim 2000$ for $\omega < 4.25$ GHz. As one goes to higher frequencies, the rise is not as great and the dielectric constant assumes a more constant value over the entire temperature range at a given frequency.

B. DISCUSSION OF ERRORS

As was pointed out in Section II, the results are obtained from measurement of the reflection coefficient, ρ, and phase, Φ, of the microwave signal. Obviously, errors in the measurement of these parameters for calibration samples and for TGS will cause errors in the final results. A fairly careful (but not complete) analysis of the effect of errors in the various measurements was made. The results of this analysis are given in what follows.

In general, $\mathcal{E}_1$ is strongly dependent on measured values of ρ and only

weakly dependent on Φ, while the reverse is true for $\mathcal{E}_2$. Also, both $\mathcal{E}_1$ and $\mathcal{E}_2$ are very sensitive to the measured parameters when the dielectric constant is large, and much less sensitive when small. Thirdly, when the dielectric constant is high, both $\mathcal{E}_1$ and $\mathcal{E}_2$ become very sensitive to Φ. These general comments apply equally to the calibration samples and to TGS.

The dependence of the results on ρ are most easy to assess: Under the worst possible circumstances a change of ρ of 0.03 changes $\mathcal{E}_1$ by 15% and $\mathcal{E}_2$ by 1%. The affect of Φ is somewhat complicated and is best explained by referring to Figure IV-1, which shows the general dependence of $\mathcal{E}$ (both $\mathcal{E}_1$ and $\mathcal{E}_2$) on Φ. One sees that a high dielectric constant means a phase near $\lambda/2$, and that in this region small errors in Φ will cause large errors in the results. The errors introduced by the various samples are discussed separately below.

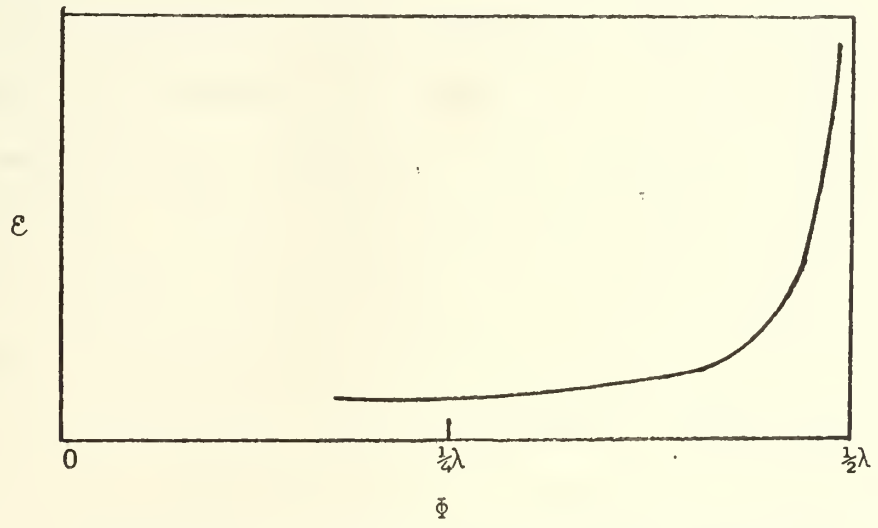

FIGURE IV-1 Behavior of dielectric constant as a function of phase.

Empty: The dielectric constant of the empty holder is 1.0 and Φ is very nearly $\lambda/4$. Thus small errors in Φ do not have much affect on the final results.

Brass: Since the phase shifts for all the samples are taken with the standing wave pattern of brass as the reference, extreme care should be

taken to insure that the brass data is as accurate as possible. It was noted that the positions of the brass minima over many runs were consistent to within ± 0.1 centimeters.

Rochelle salt: The calibration data taken for the rochelle salt sample was probably the biggest single source of error in this experiment. Φ is approximately 0.4λ when the value of the dielectric constant for rochelle salt is in the vicinity of 100. (The values for the dielectric constant for rochelle salt are taken from reference 5.) Because of this, the final results depended in a very sensitive manner on the phase for this sample. The dielectric constant for rochelle salt is temperature dependent, and pressure dependent as well since it is a piezoelectric. This in turn means that its reflection coefficient and phase are temperature and pressure dependent, thus making it difficult to get a consistent set of calibration data for the rochelle salt sample. Imperfections in the crystalline structure and the easily lost waters of hydration associated with rochelle salt also contribute to the difficulty in obtaining consistent calibration data. Finally, small variations in the value used for the dielectric constant for the rochelle salt sample also leads to small variations in the calculated value of $\mathcal{E}$ for the TGS sample.

TGS: As the dielectric constant changed with temperature, Φ varied from 0.4 to 0.5λ, approaching $\lambda/2$ as the temperature approached T_c. Because of this the final results have the largest uncertainty near T_c. The data taken for various TGS samples over the same frequency and temperature range were fairly consistent. The small variations in the measured parameters for the TGS sample are probably due to variations in material quality from sample to sample. Also, since TGS is a piezoelectric, variations in the amount of pressure applied to the sample while it was in the sample holder undoubtedly contributed to some of the variation in the data.

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There is one additional difficulty that occurs near T_c when Φ approaches $\lambda/2$ for TGS. For this value of Φ the imaginary part of the complex reflection coefficient goes to zero. This value appears in the denominator in the expression used for calculating the dielectric constant. The result is a computational error that makes $\mathcal{E}_2$ very large and drives $\mathcal{E}_1$ negative. This difficulty was experienced for some of the data runs but not for the reported results.

Taking into account the factors mentioned above, the reported values for the dielectric constant of TGS are considered correct to within a factor of two or less. Values near the transition temperature have the greatest uncertainty while those on either extreme of the temperature range have a much smaller uncertainty.

C. SUGGESTIONS FOR IMPROVEMENT

There are approximately twelve measured parameters used to calculate the final results. As discussed in the preceding paragraphs, some of these parameters affect the results in a very sensitive way. To help decrease the magnitude of uncertainty in $\mathcal{E}$, all measurements should be made as precisely as possible and, in addition, a number of data runs should be made with the different samples.

To improve on the consistency of the data, the following ideas are suggested:

1. A material other than rochelle salt should be used as a calibration standard. Ideally, the calibration standard should have a large dielectric constant similar to that for rochelle salt, but its dielectric constant should be independent of temperature and strain, and its structure should be fairly free of imperfections.

2. Minor modifications in the way the samples are prepared might prove beneficial, especially if more precision could be gained in insuring

that all samples are of uniform size and shape.

3. When working with piezoelectric material, a device that indicates how much pressure is being put on the sample by the sample holder could be used to help insure that the same amount of pressure is applied each time. It might be possible to incorporate such a device as part of the sample holder.

Suggestions for improving the data reduction procedure are:

1. The data from a number of measurements should be treated in a statistical manner. A single set of data has too large an uncertainty associated with it to yield reliable results but this uncertainty could be reduced by using averaging techniques for several sets of data.

2. The computer program used to perform the calculations should be revised so that the value of $\mathcal{E}$ is not so sensitive to small changes in the phase shift in the vicinity of the transition temperature. If this is not possible, then a larger computer with more significant figures might help to reduce the uncertainty.

It is again emphasized that the measurements made on a particular sample are characteristic of that sample, however due to the state of the art of crystal growing, defects are present in the crystal structure of the material used to make the samples, thus no two samples will be identical. This makes it difficult to get consistent data using different samples. A consistency of $\pm$ 10% is probably a realistic, though difficult, goal to strive for, given this situation.

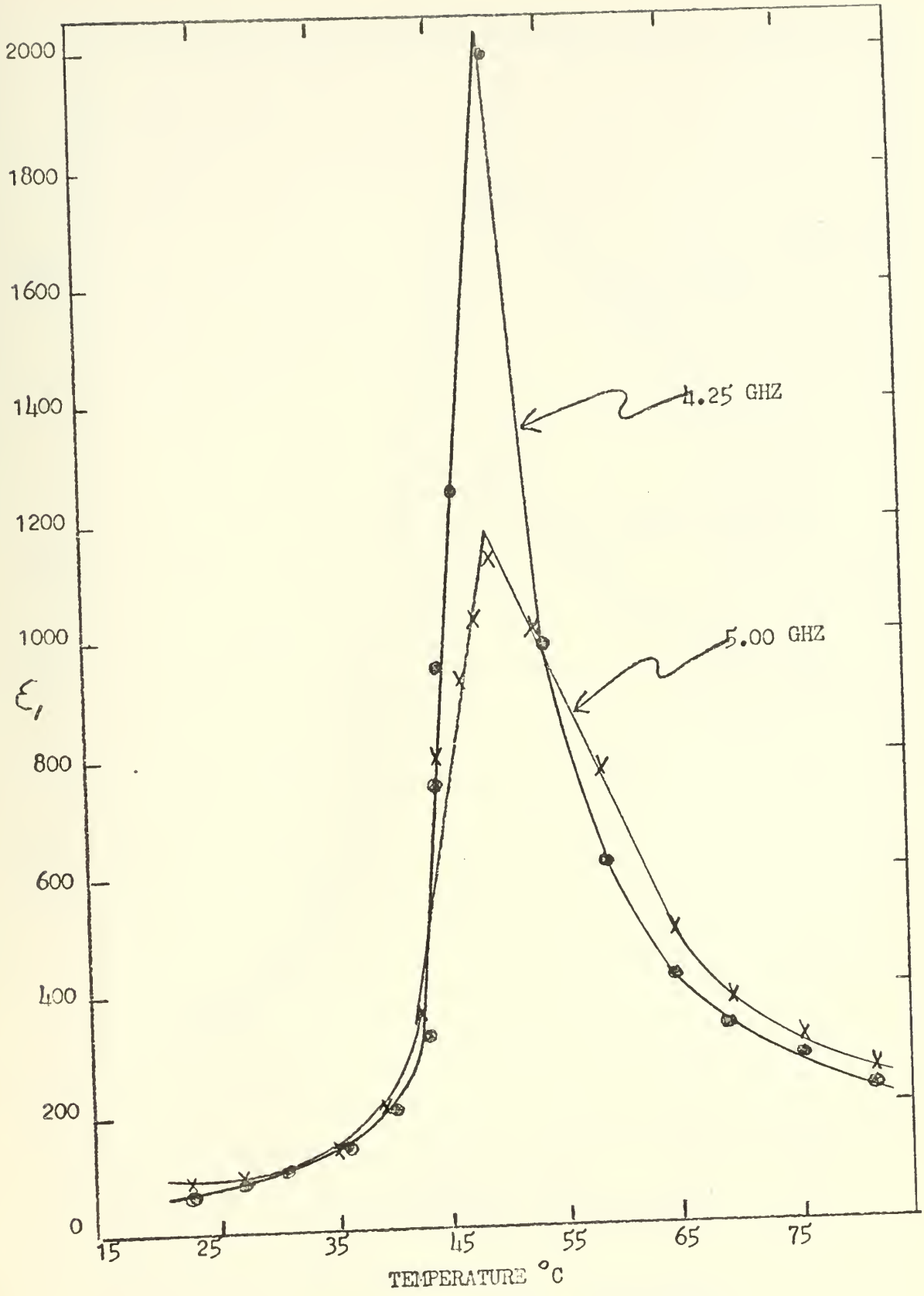

FIGURE IV - 2 DIELECTRIC CONSTANT vs TEMPERATURE

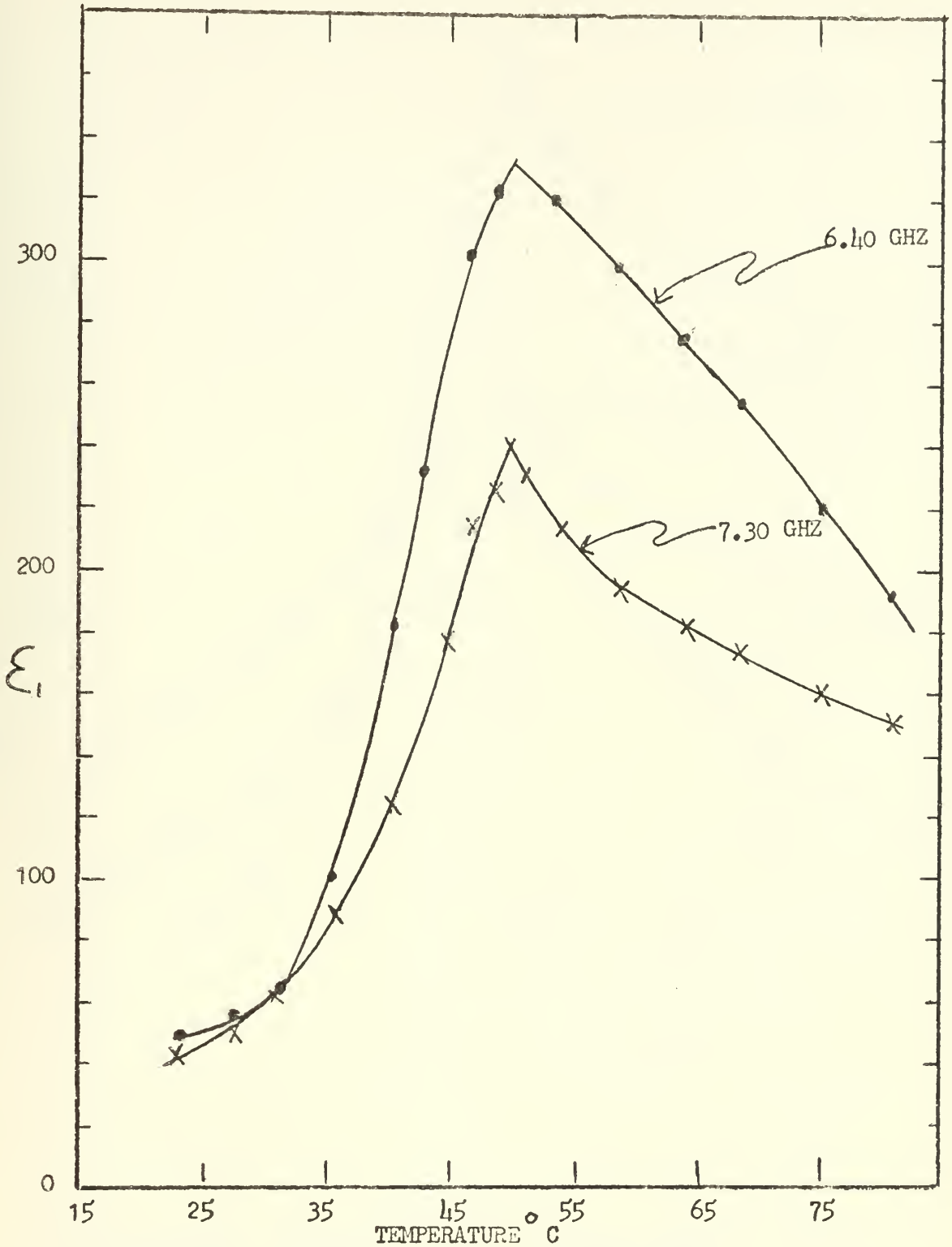

FIGURE IV-3 DIELECTRIC CONSTANT vs TEMPERATURE

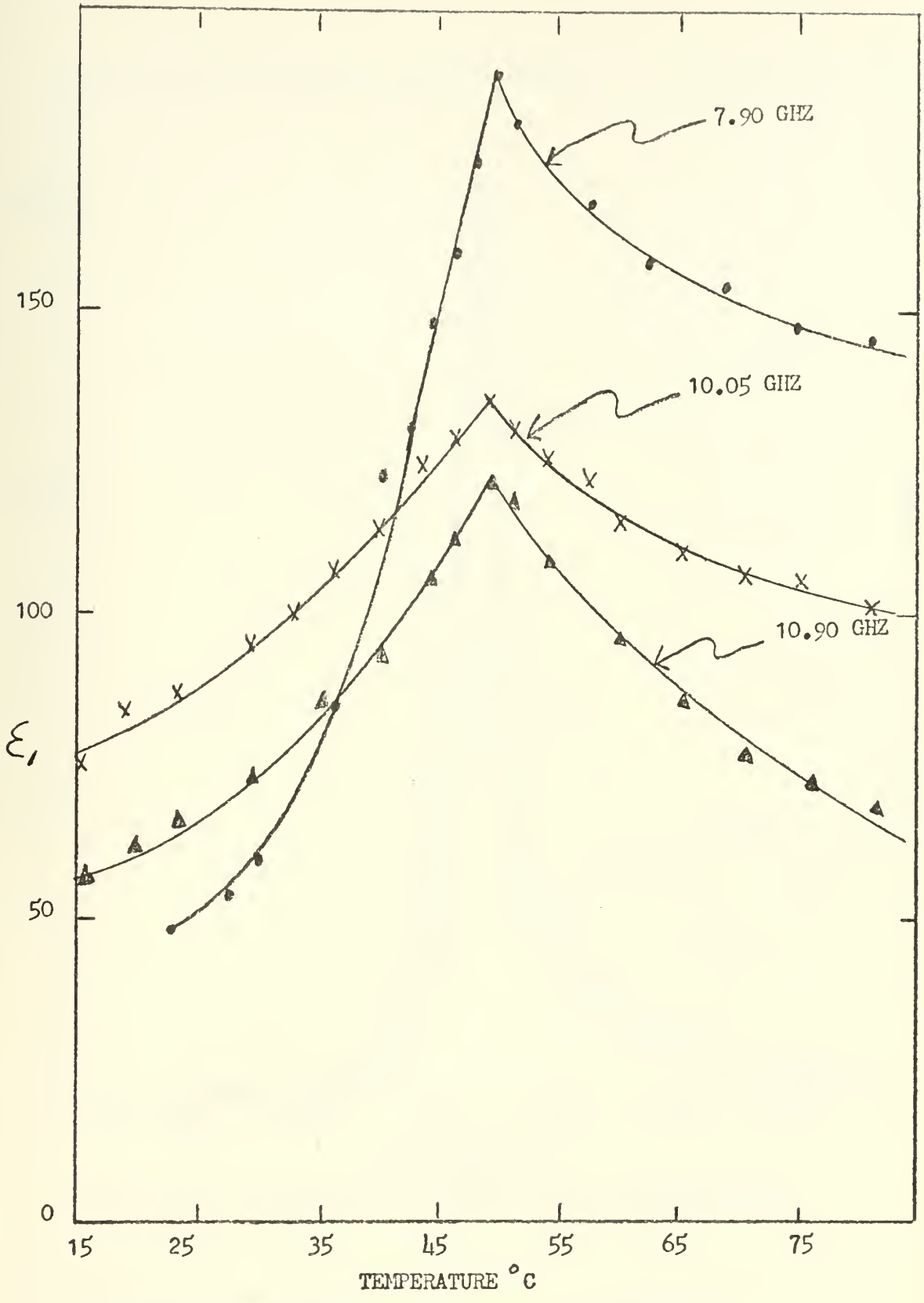

FIGURE IV - 4 DIELECTRIC CONSTANT vs TEMPERATURE

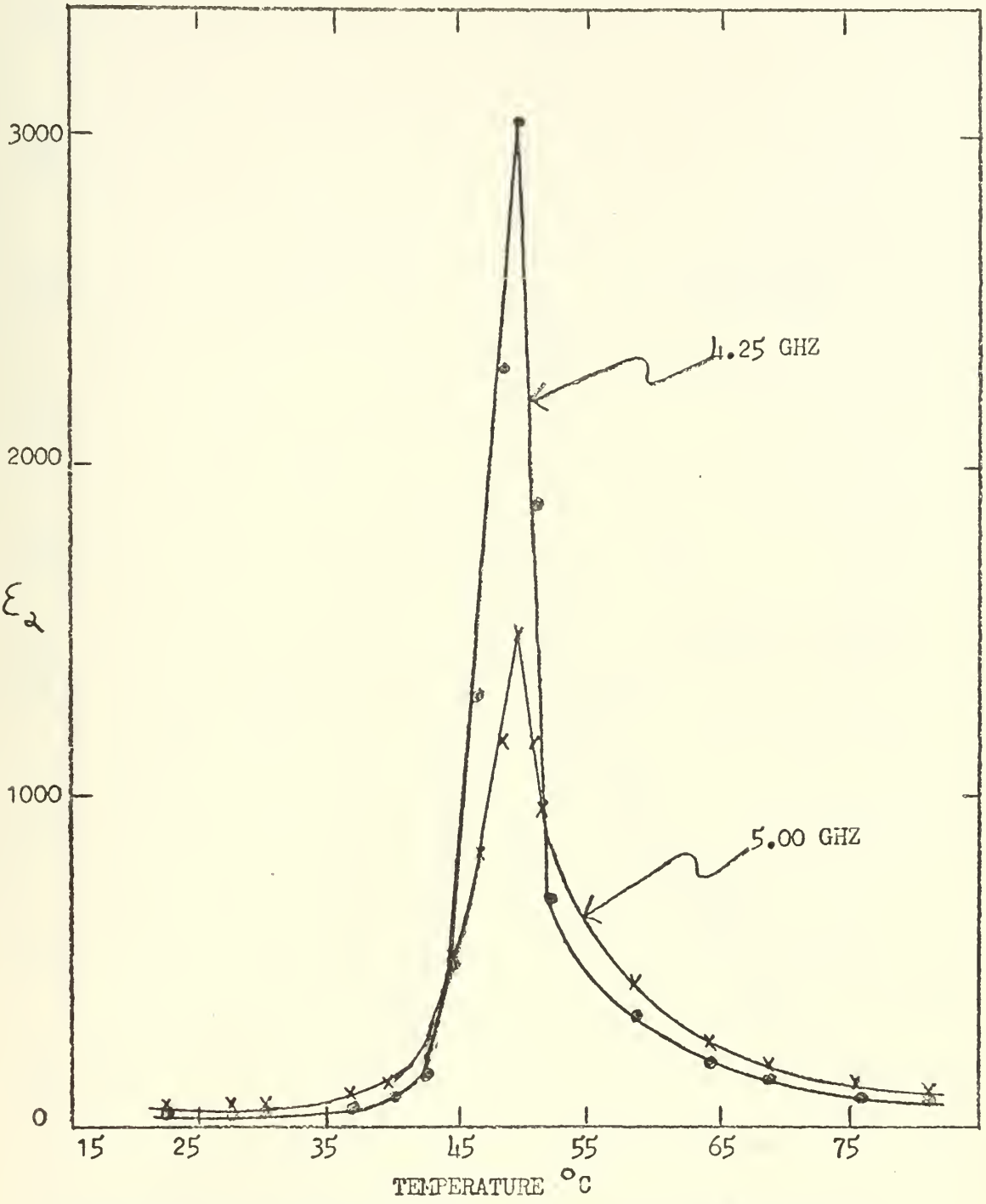

FIGURE IV - 5 DIELECTRIC CONSTANT (IMG.) vs TEMPERATURE

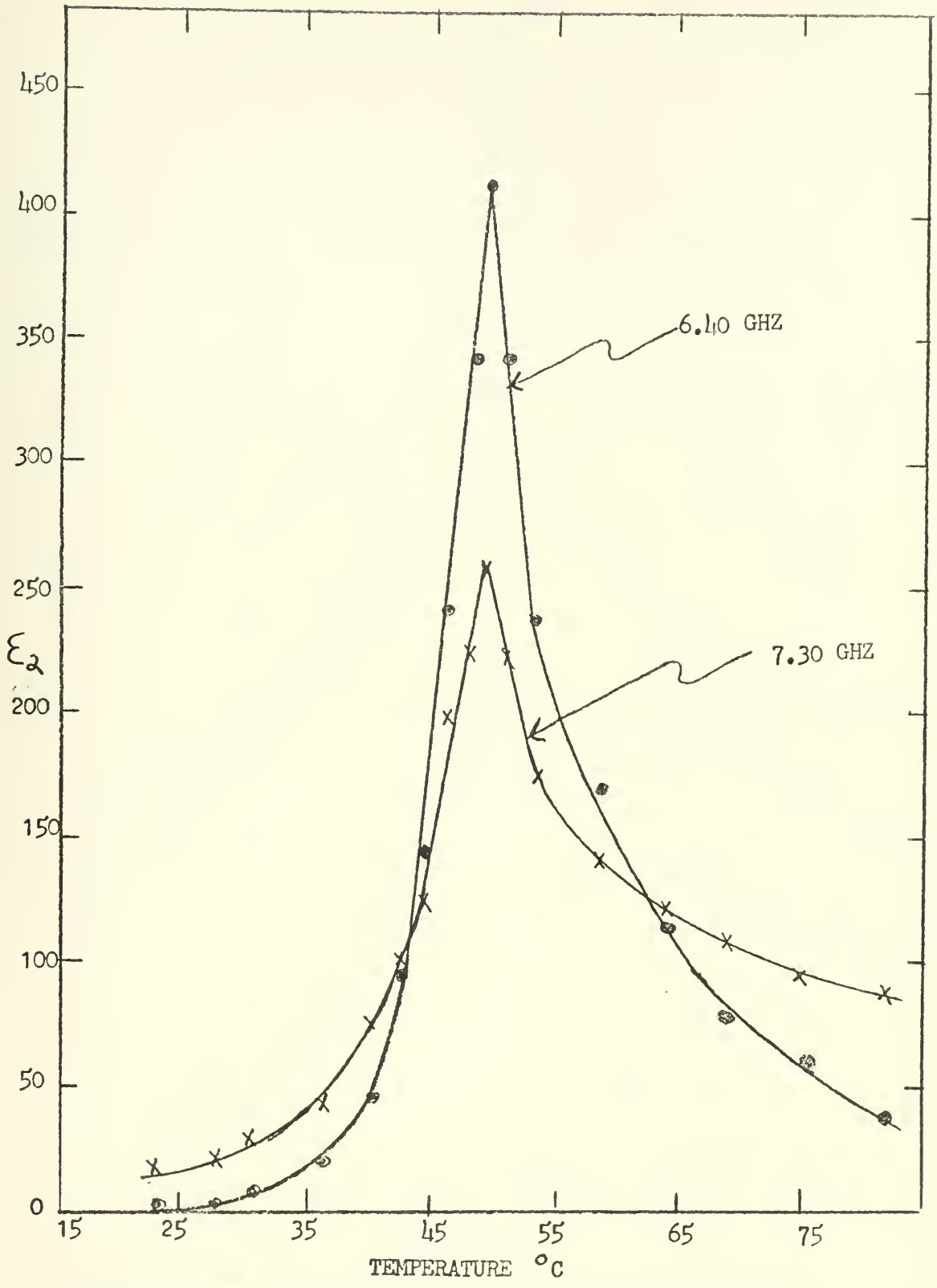

FIGURE IV - 6 DIELECTRIC CONSTANT (IMG) vs TEMPERATURE

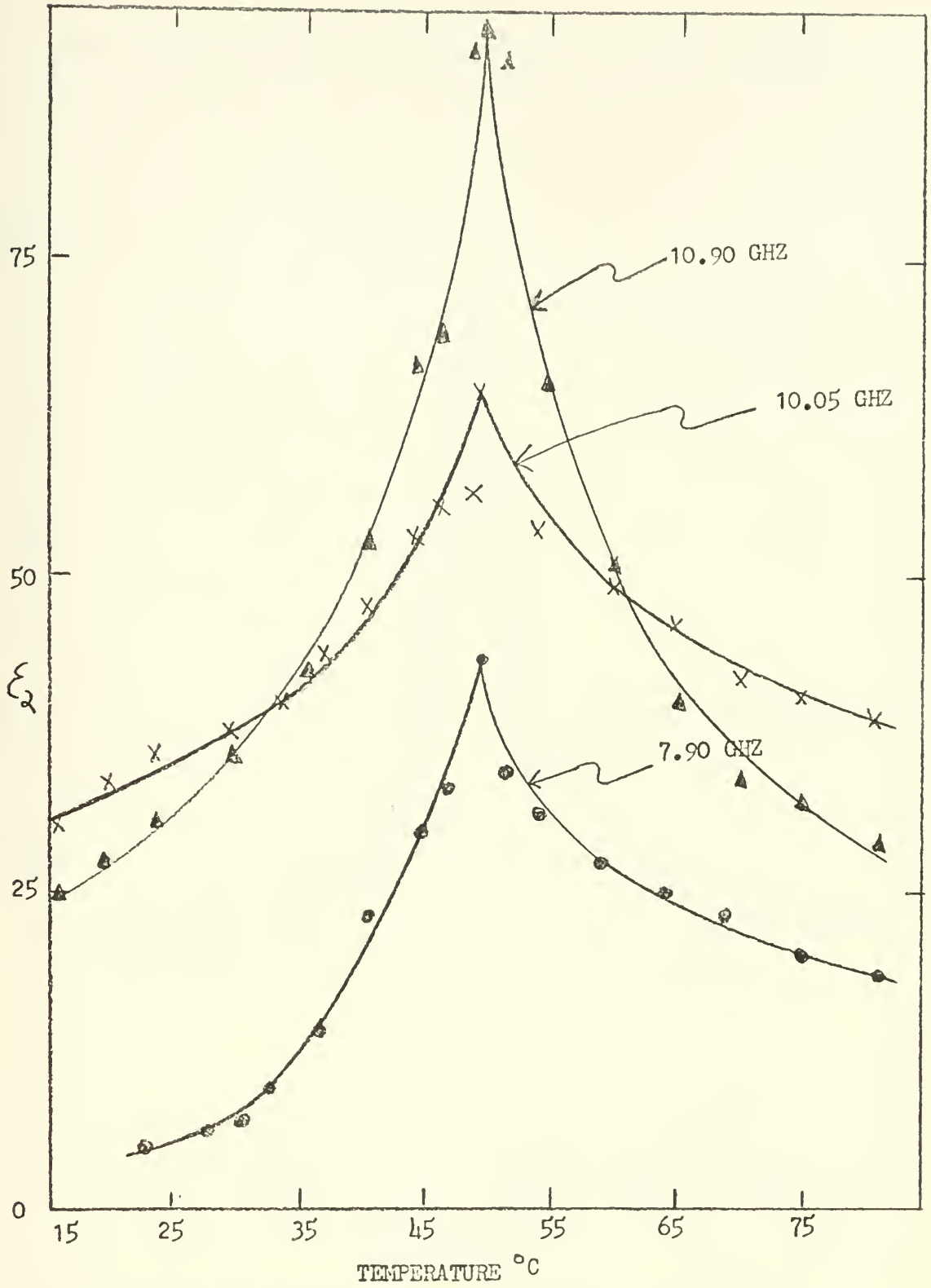

FIGURE IV - 7 DIELECTRIC CONSTANT (IMG.) vs TEMPERATURE

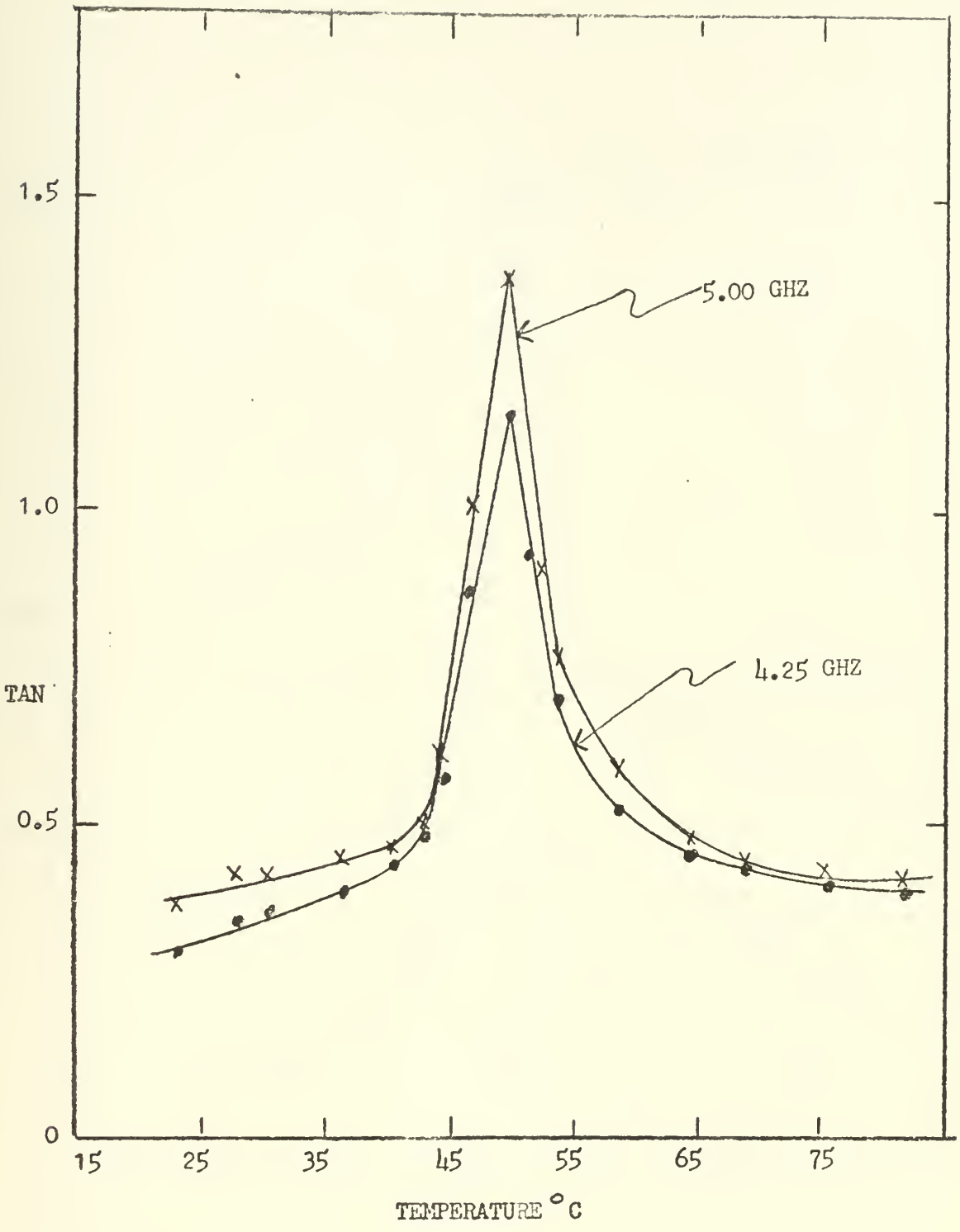

FIGURE IV - 8 LOSS TANGENT vs TEMPERATURE

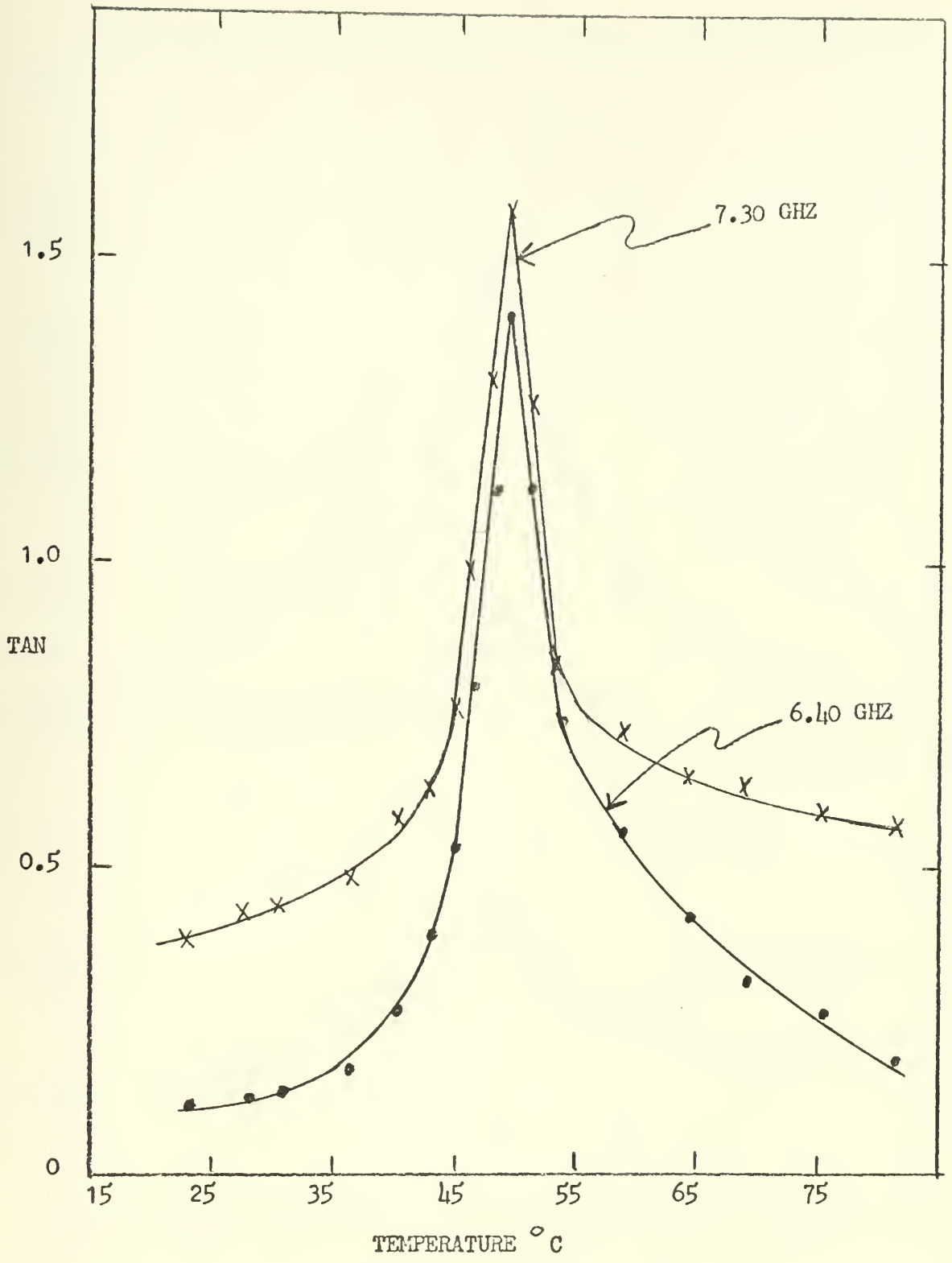

FIGURE IV - 9 LOSS TANGENT vs TEMPERATURE

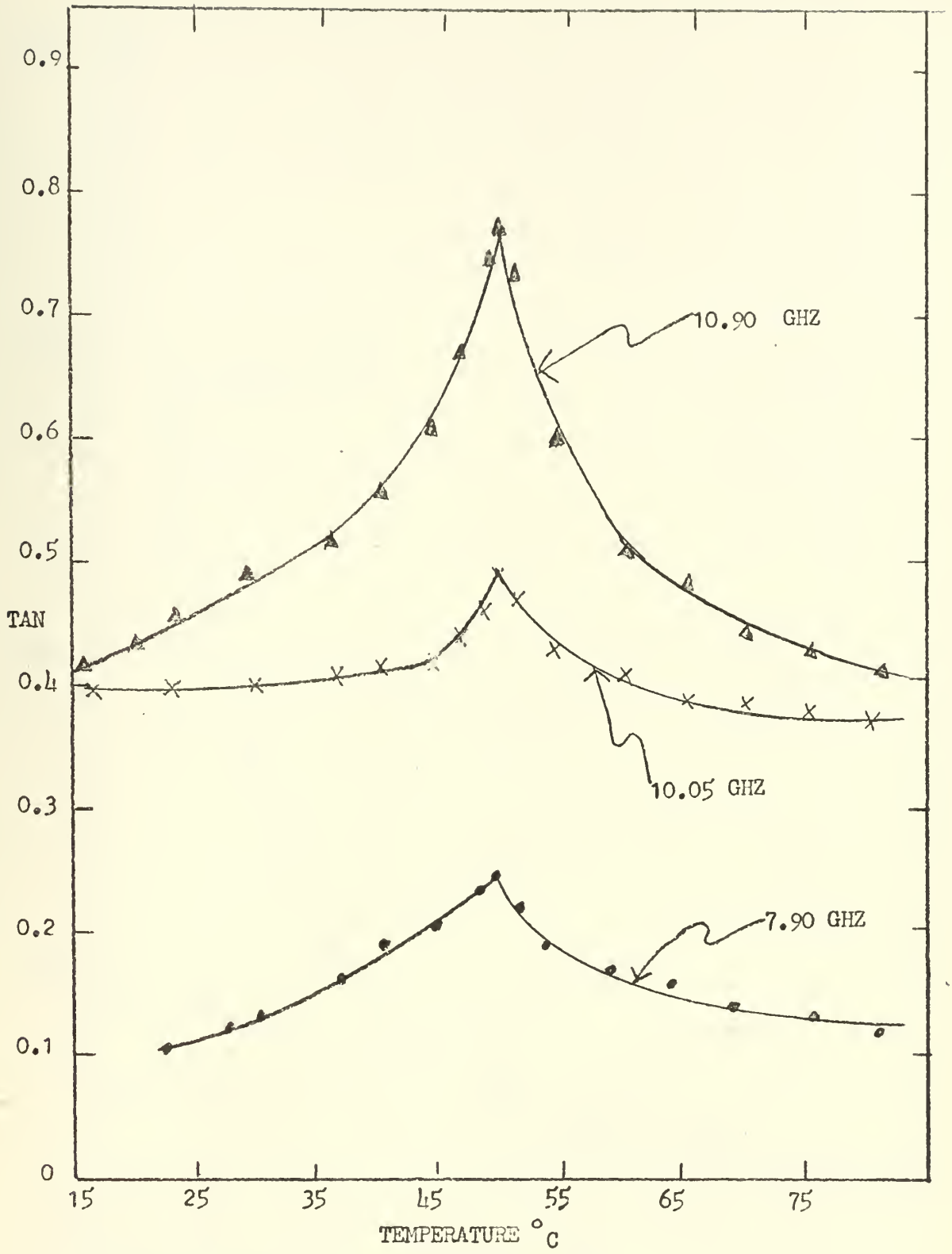

FIGURE IV - 10 LOSS TANGENT vs TEMPERATURE

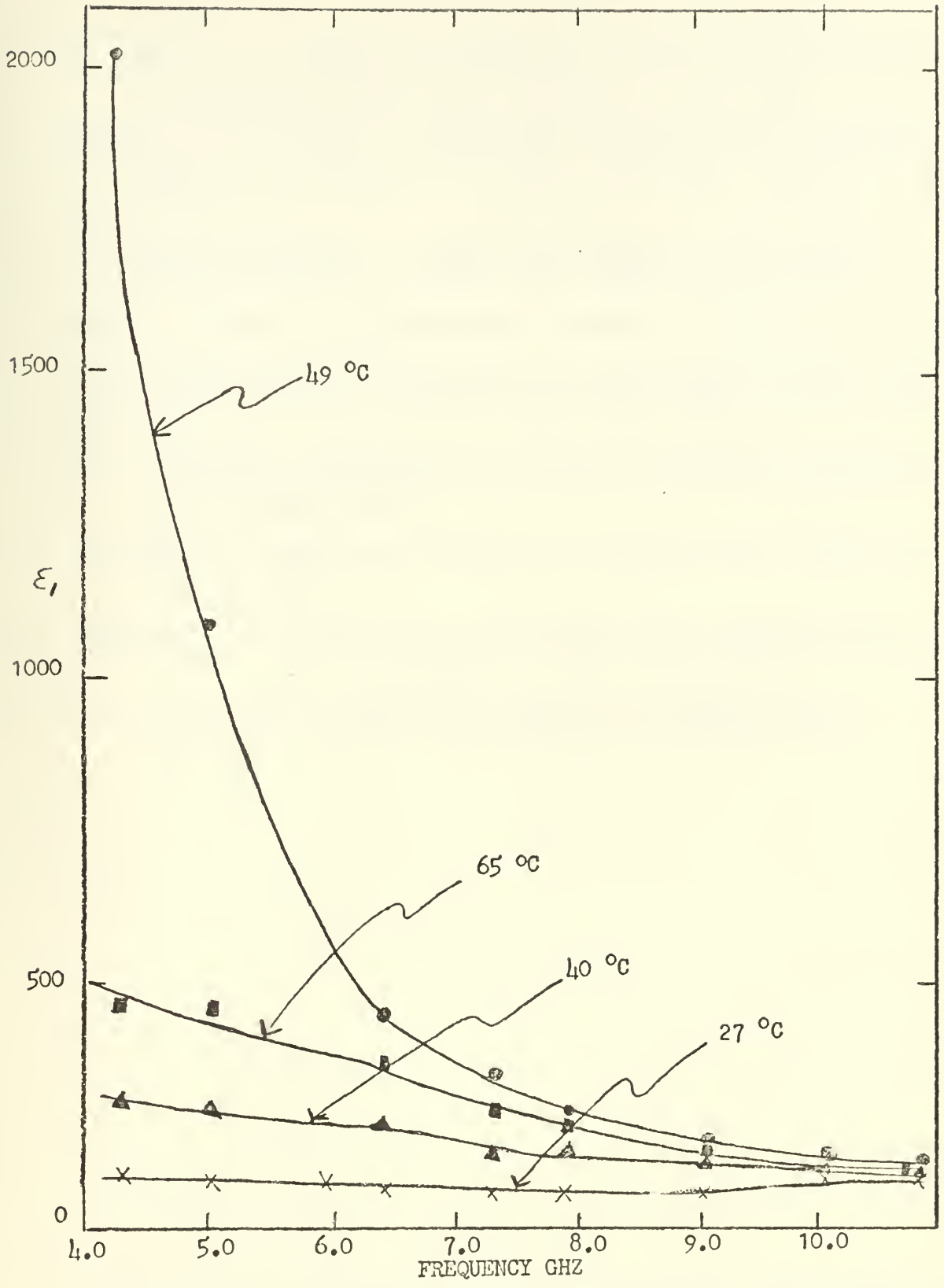

FIGURE IV - 11 DIELECTRIC CONSTANT vs FREQUENCY at FIXED TEMPERATURE

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13. ABSTRACT

This thesis describes the construction and operation of a microwave system used to measure very high dielectric constants over wide frequency and temperature ranges. Measurements of the dielectric constant of the ferroelectric triglycine sulfate are reported over a frequency range of 4.0 to 12.0 GHz. The results agree fairly well with those reported by other investigators. Sources of errors and possible improvements in this experiment are discussed.

KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
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Microwave Dielectric Properties						
Triglycine Sulfate						
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